

# Summary of work and recommendations by the “Tritium Impact” working group

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## 1 Background to the group’s work

Tritium can be naturally occurring or anthropogenic. It is produced in the form of tritiated hydrogen, tritiated water or tritiated organic molecules. Oxidation converts tritium gas to tritiated water which then joins the water cycle. Since hydrogen is a major constituent element in living matter (along with carbon, oxygen and nitrogen), the tritium can be taken up into organic molecules within cells through processes such as photosynthesis or, in animals, the biosynthesis of molecules in cells or by hydrogen exchanges with the surrounding environment. The differences in bonding forces with the organic matter lead to the definition of two sub-fractions of organically bound tritium (OBT): exchangeable OBT (exchangeable with hydrogen in cell water) and non-exchangeable OBT (more specifically, tritium bound to carbon). The exchange processes can be slowed or even stopped after the death of an organism, causing the organic molecules to remain for a certain time in the soil or sediments. Tritium is a low-energy beta emitter and is generally considered to be an element with low radiotoxicity. Tritium ingested in organic form in food is approximately three times more radiotoxic than tritiated water (dose coefficient per unit of activity ingested is approximately three times higher). This is related to the biological (elimination) half-life.

Tritium has been making the news again over the last few years. Firstly, high concentrations of tritium in organic forms have been unexpectedly observed in some marine species (flat fish, crustaceans and molluscs) in Cardiff Bay, an area which has seen industrial discharge of tritium-marked biological molecules. Analogous observations, but somewhat less marked, were also found off Sellafield, an area in which industrial discharges should theoretically be limited to tritiated water. These observations raise the issue of potential tritium accumulation along the marine food chain. Secondly, from a health perspective, recent summary reports (AGIR<sup>1</sup>, Article 31<sup>2</sup>) have highlighted various difficulties and/or uncertainties in assessing the effects of tritium exposure. Issues include the consequences of the highly heterogeneous dose distribution delivered by tritium, particularly when incorporated into DNA or histones, the uncertainties related to quality factors and the RBE (relative biological effectiveness), the value of tritium weighting factor  $W_R$  (a proposal has been made to increase the factor to 2), the lack of data on the effects of chronic exposure or the spread of results when dealing with tritiated organic molecules, with significant variation depending on molecule type and the biological effect analysed.

The ASN’s aim in setting up this working group was to examine current knowledge on all these issues.

## 2 The issue of bioaccumulation

### 2.1 Semantic clarification

As is often the case in meetings, regardless of the make-up and expertise of the groups, many discussions stem from the fact that different people understand words differently. It was important, therefore, to agree on the meaning of the terms used.

Bioconcentration means the presence of substances in an organism (e.g. aquatic organism) at a higher or lower concentration than the concentration measured in its environment (e.g. water) at the same time. The bioconcentration factor is simply the ratio between contaminant concentration in the living organism (or one of its organs or tissues) and the concentration of the same substance in the organism’s environment. Bioconcentration factors may thus be greater than, equal to or less than 1. Since bioconcentration factors are often defined in the laboratory, they *do not* take into consideration transmission up the trophic levels of the food chain. Some writers, including the authors of this report, use the terms bioconcentration and bioconcentration factor with a general and purely descriptive meaning to refer to the *increased* tritium

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<sup>1</sup> Health Protection Agency, Review of Risks from Tritium, Report of the Independent Advisory Group on Ionizing Radiation, November 2007

<sup>2</sup> European Commission, EU Scientific Seminar 2007 “Emerging Issues on Tritium and Low Energy Beta Emitters, Radiation Protection No 152, Luxembourg, 2008

concentrations observed in living organisms than in their environment, without in any way indicating the exact nature of the contamination source, the time at which such contamination occurred or the underlying biological mechanisms. Due prudence should be taken in interpreting the concentration factors.

The term *bioaccumulation* is often used with the same general meaning as bioconcentration. The French General commission for terminology and neologisms<sup>3</sup> describes bioaccumulation as a “process by which a pollutant present in a biotope penetrates or accumulates in some or all of a living organism and can become harmful; by extension, the result of this process.” In the strict sense of the word, bioaccumulation results from the gradual accumulation of a contaminant or toxic substance in an organism, from *diverse sources, including* the atmosphere, water and *food*, until an equilibrium is reached between accumulation and excretion of the substance<sup>4</sup>, with a concentration in the organism that is greater than that in the contamination sources. If this phenomenon is reproduced at each trophic level (with a cumulative increase in concentration of the substance as it moves up the food chain), the term used is *bioamplification*. The opposite process leads to dilution.

Finally, if the living organism was exposed to higher environmental concentrations in the past than in the present, said past contamination may remain in some of its tissue (*remanence*), leading to an apparent imbalance with regard to the present lower concentrations of the contaminant in the environment. This phenomenon occurs when a substance fixes in the organism at a concentration that is lower than or equal to that of the original source, and stays there for a period whose length will depend on the speed at which the substance is excreted. Remanence can also be observed in abiotic environments (soils or sediments) when physico-chemical processes cause a substance to be fixed in the environment long term.

## 2.2 Observations and interpretations

The presentations focused on observations carried out both in marine environments (Cardiff Bay, Sellafield, La Hague) and in terrestrial environments.

- In the case of the plant producing labelled molecules in Cardiff, a clear example of tritium bioconcentration was observed. The hypothesis concerning bioaccumulation/bioamplification applies if the tritium comes from tritiated organic molecules.

The tritium content found as free tritium (HTO) and organically bound tritium (OBT) in marine fauna varies by factors between 1,000 and 10,000 with respect to the concentrations found as HTO in the sea water

- For the Sellafield site (fuel reprocessing facility managed by BNGSL British Nuclear Group Sellafield Limited, which discharges tritiated water), the tritium content found as free tritium (HTO) or organically bound tritium (OBT) in marine fauna (fish, crustaceans and molluscs) varies by a factor of 10 with respect to the concentrations found as HTO in the sea water with 1 to 2 years hysteresis (time lag) between the maximum discharge values and the maximum tritium values in molluscs and flat fish. These observations were interpreted in different ways by various members of the working group.

Some consider that the abnormally high concentrations measured in fish near Sellafield may result either from remanence in sediment labelling following previously large discharges or from the existence of tritiated organic molecules in the same water outflow.

Others consider that this is a case of bioaccumulation which is related to the discharge of *tritiated water*. According to proponents of this view, the hypothesis of marine currents carrying tritium-labelled organic molecules discharged by the Cardiff radiochemical plant is refuted by the fact that analysis carried out near the Wylfa nuclear power plant (NPP), on the west coast of the UK *between* Cardiff and Sellafield did not find any detectable presence of tritium in the marine fauna.

<sup>3</sup> Journal Officiel de la République française, Avis et Communications, 4 February 2010

<sup>4</sup> IRSN, “Le tritium dans l’environnement”, Report DEI 2009-05

Other members of the group felt they did not have enough information to form a view.

- For the La Hague site (fuel reprocessing facility run by AREVA NC, discharging tritiated water), the Institute for Radiological Protection and Nuclear Safety (IRSN) considers that the research into tritium in the environment around La Hague (joint IRSN-AREVA campaigns) do not show any tritium bioaccumulation / bioconcentration. It should be noted that values for tritium content found as HTO in the sea water around the Sellafield outflows in the Irish Sea and around the La Hague outflows in the Channel are substantially similar.

The French Association for the control of radioactivity in the West (ACRO) reports that bioconcentration (by a factor between 2 and 7) was found in an older joint EDF-IRSN study (from the period 1981-1985) in a small number of available mollusc, crustacean and fish samples. ARCO highlights the fact that measurements performed on marine organisms in Cardiff Bay suggest that algae are a poor indicator for monitoring tritium contamination in marine fauna, since tritium concentrations were significantly higher (by a factor of over 10) in fish than in algae. Its position is that the choices made by IRSN and AREVA to take measurements in algae for monitoring purposes is not the most appropriate method for identifying concentration phenomena in marine organisms. ACRO also points to the very limited number of measurements taken around La Hague between 2000 and 2009 on edible marine produce (flat fish, crustaceans, molluscs etc.).

- With regard to the terrestrial environment, the French Atomic Energy Commission (CEA) presented the mechanisms by which tritiated water is incorporated into plants, based on work performed by the International Atomic Energy Agency. The CEA also reported the results of monitoring measurements performed at the Valduc site by the CEA and, in parallel but independently, by SEIVA, a local association. The levels of radioactivity in water produced in combustion of organic plant matter were found to be between the activity in airborne water vapour and the activity in groundwater. In the CEA's view, these results do not reveal any phenomenon of concentration in the organic matter. According to the CEA, the IAEA calculations could be excessively conservative, by overestimating the free water concentration in plants.
- Finally, high concentrations of organic tritium have been measured in the sediment of water courses affected by discharges from the watchmaking industry, with the organically bound tritium (OBT) content varying with respect to HTO content in the river water by factors of between 1,000 and 10,000.

**By way of conclusion, the group considers that the only way to clear up doubts, assess the effect of the various factors at stake, in particular regarding the distribution of tritium in the different compartments (including organic matter in sediment) and to better define the free and organically-bound tritium components in living species is to use appropriate environmental measurement campaigns, with a scientific approach. In the marine environment, these campaigns must focus on a large enough number of edible marine produce samples from various trophic levels (flat fish, crustaceans, molluscs etc.).**

Some old publications drawing on environmental data (from the 1970s and 1980s) suggest that tritium could bioamplify in some aquatic trophic chains and that the nutritional pathway is predominant with respect to the direct pathway (water). Research has since become rather rare on this subject.

International studies were recently analysed in a summary report by the IAEA (EMRAS programme 2006-2009) and a calculation model has been proposed (TRS No. 472 and Tecdoc No. 1616). These models now explicitly take into account tritiated organic matter formed from tritiated water (by tritium-labelling food), leading to no increase in concentration at higher trophic levels. However this does not prejudice any transfers and incorporation following the discharge of specific tritiated organic molecules.

Summary report IRSN DEI 2009-05 states that “on the basis of currently available knowledge and with “normal” environmental activities, no phenomenon has been identified as liable to cause significant “bioaccumulation” over the long term and no measurement has pointed to this.” The same report does however mention that “for animal organisms, there is *little data* with respect to the complexity of the issue (number of processes involved, interactions and variability according to species, age and diet)”.

**The group wishes to highlight the still fragmentary nature of current knowledge on remanence and on tritium behaviour in sediment, and the need to use targeted multidisciplinary studies with rigorous protocols to provide experimental verification of the hypotheses put forward in older studies, in particular regarding the possible influence of the activity of microorganisms in aquatic sediments when organic tritium is remobilised in aquatic animal organisms. In general, the scientific data regarding the conversion of tritiated water into organic tritium along the food chain should be enhanced. Reliable quantitative estimates are required.**

### 3 Measurements

In discussing tritium, it is essential to state what form it is in (HTO, HT, OBT) and when talking about OBT, it must be clearly specified whether we are talking about the food chain or a molecule that has been labelled for research purposes.

One of the reasons for divergences in the measurement results and in the interpretation thereof comes from difficulties related to metrology and the representative nature of measurements and the lack of a standardised protocol. Methodological clarification is required in order to be able to *compare* environmental tritium measurements.

The *Commission d'ETAbblissement des Méthodes d'Analyse* (CETAMA), a French working group whose role is to improve the quality of measurements by organising collaborations between laboratories, has done some review work on tritium measurements. In practice, routine environmental monitoring in France currently only focuses on measurements of free tritium (HTO). CETAMA is now working to validate measurement methods for total organically bound tritium (intercomparison exercise 2009-2010). Measurements of non-exchangeable OBT (NE-OBT) are unlikely to be validated until at least 5 years from now. This type of routine measurement still raises metrological problems (the E-OBT and NE-OBT separation test is still unreliable, and analysis takes several days) and unanimous agreement has not been reached. In the opinion of CETAMA, further research is required to improve this measurement in order to better understand the various transfer factors in the environment.

**The working group agreed on the need to continue this work to validate and standardise measurement and sampling methods and protocols (“to be sure of what is measured”), and that this work should be carried out within an international framework.**

#### 3.1 Nature of releases

There is another open question – are there sources of tritium releases in organic form other than those from industries that synthetically create labelled molecules?

The aforementioned IRSN report highlights the gaps that exist in knowledge regarding the presence of labelled molecules with high levels of specific activity, their behaviour and the consequences in terms of tritium accumulation. The IRSN feels that a key priority is to carry out metrology studies into the various physico-chemical forms (speciation) in which organic tritium is liable to affect humans.

For its part, AREVA considers that after more than 10 years operating with solvent in its plants, no tritium transfer has been observed from tritiated zones to non-tritiated zones via the solvent. It further concludes, on the basis of concentration factors observed in algae, molluscs, crustaceans and fish, that the chemical form of tritium discharges is principally HTO.

**Given the major diversity of tritiated organic molecules, the group agrees on the need for caution in conclusions and extrapolations and on the need to characterise the chemical speciation of discharges from potentially relevant sites.**

## 4 Health effects of tritium

### 4.1 Relevance of “mean organ dose”

Should the effects of beta radiation from tritium be reassessed?

This question is raised by the fact that the tritium isotope has certain specific features. The electron path is very short (less than the diameter of a cell and even of a cell nucleus) and the ionisation density is high, which can cause cluster damage to DNA if the tritiated molecules get into the cell nucleus. Two further phenomena contribute to locally enhancing the effects of tritium – its *in situ* transmutation into helium and enrichment of water in the DNA hydration shell, converting it to tritiated water (referred to as “buried tritium” or the “isotope effect”). All these physico-chemical effects cause lesions, which can in turn lead to DNA mutations. Although the dose distribution is relatively homogenous when the tritium takes the form of tritiated water, it is *heterogeneous* when it is incorporated in DNA or histones. The issue of the relevance of the *mean organ dose* concept as a risk indicator therefore arises. In other words, doses calculated according to the conventional method (using the ICRP’s Sv/Bq conversion factors) could lead to an incorrect estimate of the risk.

In comparing the studies on the biological and health effects, it would be useful to harmonise the methods used to estimate the dose at different scales (cells and organs), according to the form of the tritium, the exposure pathway, the length of exposure and the time before analysis.

Further research into the biological effectiveness of tritium, in particular during various stages of pregnancy, is required in order to clear up these uncertainties and gaps in knowledge.

In recent report<sup>5</sup>, the IRSN stated that “the dosimetric approach to risk is deemed to be robust and lists tritium as one of the least radiotoxic radionuclides,” but nevertheless suggested that “data is lacking on the metabolism and biological effects associated with organic tritium in situations of environmental exposure” and that experimental radio-biological studies should be carried out on various forms of OBT, using a specially adapted technical set-up, in the context of Europe-wide cooperation.

**The group agrees on the need to use up-to-date methods to gather further knowledge on the effects of tritium, in order to comprehensively characterise the physico-chemical forms used (covering a concentration scale that would include industrial discharges) and the biological mechanisms at work, focusing not only on aspects relating to carcinogenesis, and looking at age at the time of exposure and at differences between accident-related and chronic exposure.**

### 4.2 $W_R$ and RBE

For reference, the radiation weighting factor ( $W_R$ ) is used in health physics to take into account the effect of radiation *type* in inducing long-term stochastic effects such as *cancer* or hereditary effects. Opinions were divided within the group with regard to the need to increase the value for tritium (currently 1).

In opposition to the findings of the 2007 AGIR report in the UK and the EURATOM treaty “Article 31 experts” (cf. 2007 Scientific Seminar), the ICRP recently confirmed its choice of a radiation weighting factor of 1 for tritium and low-energy beta emitters, taking into account on the one hand the uncertainties around the issue and on the other hand, purely *forward-looking* objectives for the radiation protection system and the priority to be placed on optimisation and dose constraints.

The IRSN is of the opinion that the RBE of tritium for stochastic effects, on which the weighting factor  $W_R$  is based, is closer to 2 than 1, but considers that choosing a weighting factor  $W_R$  of 2 rather than 1 would only have a minor significance in routine situations and should only be used in assessing individual risks. This opinion is not shared by the associations ACRO and ANCLI, which are arguing for a weighting factor of 5 for the sake of precaution. No consensus was reached on this issue within the group.

### 4.3 Epidemiological studies

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<sup>5</sup> IRSN, “Eléments de réflexion sur le risque sanitaire posé par le tritium”, 2009

A literature review shows that studies on exposed workers reveal that risk of cancer is not increased. However, the studies are limited in their robustness due to the insufficient statistical power and/or lack of information on tritium dose. It should be noted that the tritium doses logged were of the order of ten mSv, which implies that very large cohorts would be needed to demonstrate a statistically significant difference between the exposed group and control group. A coordinated international approach based on standardised dosimetric assessments would be required in order to make progress in this field. Within France, a vital first step would be to set up a tritium database to enable tritium to be taken into account in epidemiological studies.

Furthermore, there have been very few studies into the effects of tritium on the population. The studies that do exist are geographical and not very informative. In general terms, international multi-centre studies would be the only potential way to achieve statistical power that is large enough to offer a relevant response to the epidemiological questions. For populations living close to nuclear facilities, the issue of statistical power in the epidemiological studies is more acute when the doses are lower. The issue of the epidemiological *detectability* of tritium risk is raised and this highlights the potentially relevant role that molecular studies with biomarkers could play. Currently, the existing epidemiological studies on tritium in populations have been shown to be of no practical relevance. This does not mean that epidemiological monitoring is unnecessary – it is essential, as for any site posing an industrial hazard.

**The group agrees on the importance of assessing the *feasibility* of epidemiological studies in French workers, since it would be useful to gather data on tritium exposure and to process this data in a coordinated manner alongside other ongoing studies around the world.**

#### 4.4 Hereditary effects

No increase in hereditary effects have been observed to date in the human race, either in the descendants of people exposed to radiations (Hiroshima and Nagasaki survivors; radiation-treated patients or workers exposed to ionising radiation) or in regions with high natural background radioactivity.

Quantitative risk assessments are therefore based on indirect data – firstly, on the frequency of spontaneous mutations in the germline of the human race and secondly, on experimental studies tracking the descendants of radiation-exposed rodents.

There is very little *tritium-specific* data and the risk assessments are taken from calculations based on data on the hereditary effects of ionising radiation in general.

Thus, based on the fact that there is no cell division in oocytes from the foetal period until fertilisation, British researchers calculated the risk of a woman passing on a radiation-induced anomaly 30 years after her mother ate Cardiff Bay fish containing OBT during pregnancy. The oocyte dose was estimated to be 2.7-5.4 mGy and the calculated risk value was an undetectable increase in risk.

The handful of experimental studies performed in the USA on induced mutations (spermatogenesis) following chronic exposure to HTO gave results that were similar to those obtained with X-ray or gamma radiation exposure. However, reservations should be expressed as to the validity of cross-species extrapolation based on these experimental studies.

**Data regarding the potential induction of hereditary effects should be critically assessed and with great care. New approaches should be investigated, in light of the latest advances in biology.** New tools in the fields of genetics and cellular imaging can be used to analyse the transmission of lesions and their consequences (by studying recessive mutations, non-coding regions that are important for verifying genome integrity, gene expression, etc.). In addition, genomics tools can be used to identify the transmission of a hereditary characteristic (not spontaneously apparent) through several generations of a family.

#### 4.5 In utero exposure

The former Chairman of German radiation protection commission SSK presented and discussed experimental data on the effects of embryo exposure to tritiated thymidine and tritiated arginine (histone

precursor) at the pre-implantation stage of pregnancy (*in vitro* experiments). Given the heterogeneous distribution and specific incorporation into DNA, tritiated thymidine is 1,000 to 5,000 times more effective than tritiated water in inducing harmful effects at the same level of applied activity. The effect is even more marked with tritiated arginine (factor of 10,000) and can be observed at lower levels of activity. Given mechanisms at work, this observation could be relevant to other cell types and systems with regard to mutagenicity. However, it should be noted that these *in vitro* studies performed at high levels of specific activities are not necessarily representative of *in vivo* situations.

The CEA (Life Sciences Department) presented a summary of work into the *in utero* effects of tritium. Tritium (HTO or tritiated organic molecules) crosses the placental barrier fairly easily. As for other ionising radiations, beta radiation from tritium causes local apoptosis and mutagenesis, which can lead to tumours or functional consequences by negatively affecting organogenesis (the mechanisms of cell proliferation, migration and differentiation that are closely linked in an embryo). The central nervous system seems to be a particularly vulnerable target. Tritium concentration in this system is 3 to 20 times higher than in other organs, and at values of just a few cGy (roughly 1 GBq.L<sup>-1</sup> of tritium), deterioration of cognitive functions can be observed, along with a reduction in the number of neural cells. Transfer of tritiated organic molecules to the foetus is a process of active transport and the molecules are preferentially incorporated into the DNA of cells that are actively multiplying. The CEA has concluded that the studies currently available are difficult to analyse because of their disparate nature and that further in-depth study is required into the effects of tritium (HTO and tritiated organic molecules) after exposure during *in utero* development.

**All members of the group agree that further research is vital in order to improve knowledge of the effects of tritium exposure on embryos and foetuses.**

### **5 Recommendations from the “Tritium Impact” working group**

The various recommendations issued by the group are summarised below. Clearly, the one key word that comes up repeatedly is (further) “research”.

With respect to the environment, the group recommends the following (in order of priority):

1. Current work to validate and standardise measurement and sampling methods and protocols (“to be sure of what is measured”) should be continued and carried out within an international framework;
2. Appropriate environmental measurement campaigns should be used, with a scientific approach to clear up doubts and assess the effect of the various factors at stake, in particular regarding the distribution of tritium in the different compartments (including organic matter in sediment) and to better define the free and organically-bound tritium components in living species. In the marine environment, these campaigns must focus on a large enough number of edible marine produce samples from various trophic levels (flat fish, crustaceans, molluscs, etc.);
3. Given the major diversity of tritiated organic molecules, caution is needed in drawing conclusions and making extrapolations and the chemical speciation of discharges from potentially relevant sites needs to be characterised;
4. Given the still fragmentary nature of current knowledge on remanence and on tritium behaviour in sediment, targeted multidisciplinary studies with rigorous protocols need to be used to provide experimental verification of the hypotheses put forward in older studies, in particular regarding the possible influence of the activity of microorganisms in aquatic sediments when organic tritium is remobilised in aquatic animal organisms. In general, the scientific data regarding the conversion of tritiated water into organic tritium along the food chain should be enhanced. Reliable quantitative estimates are required.

With respect to health effects, the group recommends the following (in priority order):

1. Up-to-date methods should be used to gather further knowledge on the effects of tritium, in order to enable comprehensive characterisation of the physico-chemical forms used (covering a concentration scale that would include industrial discharges) and the biological mechanisms at work, focusing not

only on aspects relating to carcinogenesis, and looking at age at the time of exposure and at differences between accident-related and chronic exposure;

2. Knowledge of the effects of tritium exposure on embryos and foetuses should be improved. Further research in this area is vital;
3. Data regarding the potential induction of hereditary effects should be critically assessed and with great care. New approaches should be investigated, in light of the latest advances in biology;
4. The *feasibility* of epidemiological studies in French workers should be assessed, since it would be useful to gather data on tritium exposure and to process this data in a coordinated manner alongside other ongoing studies around the world;
5. A radiation weighting factor ( $w_R$ ) of 2 (instead of 1) should be used in individual risk assessment situations. No consensus was reached within the group as to the factor to be used in routine situations.